Magnetic Phase Modulation of Recoilless Gamma Radiation by Nuclear Zeeman Effect

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Novel time-domain data on the nuclear Zeeman effect are presented from ^{67}Zn Mössbauer experiments. The results are in accordance with a theory developed for magnetic phase modulation. The self-calibrating feature of the high-frequency Zeeman scanning method allows precise determination of the 93-keV-state magnetic moment of the ^{67}Zn nucleus. For ^{57}Fe in magnetically soft materials, the present approach predicts rf-field--induced sidebands without any mechanical motion, and an associated collapse phenomenon.

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A phase-modulation theory is derived to analyze the influence of a periodically varying magnetic field on the Mössbauer resonance. Results are presented from novel Zeeman scanning experiments in which the *time dependence* of the transmission intensity is measured within the period of the rapidly varying magnetic field.[1] The use of the high-resolution Mössbauer resonance of ^{67}Zn in diamagnetic ZnO allows pure magnetic modulation to be produced without spurious mechanical vibrations.[2] Compared with ^{57}Fe, magnetic fields and modulation frequencies 3 orders of magnitude lower are sufficient.

Mössbauer velocity spectra of ferromagnetic foils exposed to strong radio-frequency (rf) magnetic fields show line broadening[3] or sidebands[4–10] depending on the frequency of the modulation relative to the experimental linewidth. In magnetically soft materials, the external rf field can produce a collapse of the static hyperfine pattern to a single resonance line.[7,8] A discussion on the origin of the sidebands observed in these experiments has persisted for almost 20 years. Mechanisms proposed include mechanical vibrations due to magnetostriction and magnetic-domain-wall motion. Considerable experimental evidence has been provided in support of magnetostrictive coupling in generating sidebands.[6]

On the basis of the phase-modulation theory, we present a new model for the Mössbauer resonance in a *periodically* switching magnetic hyperfine field, and predict both the sidebands and the associated collapse phenomenon. These effects are expected to play a dominant role in experiments with magnetically soft materials, provided that the contribution of mechanical vibrations is eliminated, e.g., by use of samples of very low magnetostriction.

We first develop a formalism needed to describe the time dependence of the transmission intensity as the Mössbauer resonance is passed in a time comparable to the lifetime of the excited nuclear state. It is assumed that an excited source nucleus emits a classical, exponentially decaying electric field which oscillates at the angular frequency ω_s corresponding to the energy of the gamma ray.[11] According to the model, mechanical motion introduces modulation of the phase of the field.[12–15] Analogously, shifting of nuclear Zeeman levels by an alternating magnetic field should generate phase changes. The phase-modulation function $\phi(t)$ can be calculated as an integral of the time-dependent Zeeman energy. As an example we consider magnetic phase modulation relevant to the ^{67}Zn experiments presented below. When the magnetic field oscillates sinusoidally, $\mathbf{B}(t)=\mathbf{B}_0\times\cos(\Omega t)$, the phase-modulation function for an excited nuclear state with spin $I_e=\frac{1}{2}$ is

$$\phi(t)=-\frac{1}{\hbar}\int_0^t g_e\mu_N\mathbf{I}_e\cdot\mathbf{B}(\tau)d\tau=\pm a\sin(\Omega t), \qquad (1)$$

where g_e is the gyromagnetic ratio, μ_N is the nuclear magneton, and $a=g_e\mu_N B_0/2\hbar\Omega$ is the amplitude of phase modulation. In general, the modulation depends also on the magnetic splitting of the ground state.

In the following, we consider separately the different components of the recoilless source radiation. With nondiagonal Hamiltonians, the ac-field–induced transitions between the nuclear sublevels are neglected. The periodically modulated part of the radiation field is expanded as

$$e^{i\phi(t)}=\sum_{n=-\infty}^{\infty}C_n(a)e^{in\Omega t}$$

in terms of the modulation frequency Ω. The Fourier coefficients $C_n(a)$ determine the sideband amplitudes and they can be calculated from $\phi(t)$ by integration over the period $T=2\pi/\Omega$ of the modulation:

$$C_n(a)=\frac{1}{T}\int_0^T d\tau\exp\{i[\phi(\tau)-n\Omega\tau]\}. \qquad (2)$$

The Fourier coefficients corresponding to the sinusoidal $\phi(t)$ of Eq. (1) are Bessel functions, $C_n(a)=J_n(a)$.

For calculation of the transmission intensity, the absorber nuclei are described by the response of a harmonic oscillator with resonance frequency ω_a. With use of the general coefficients $C_n(a)$, the time dependence of

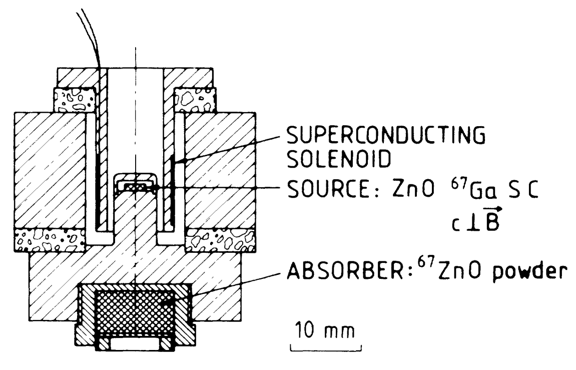

FIG. 1. Experimental setup for magnetic modulation of the ^{67}Zn Mössbauer radiation. The supporting structure has been designed to eliminate any spurious mechanical noise due to the alternating electric current of the solenoid. The inhomogeneity of the magnetic field at the active source volume is $\pm 0.5\%$. The device is operated in helium gas at a temperature of 4 K.

the transmission intensity is given by[13,15]

$$I(t;\Delta\omega)=\sum_{n=-\infty}^{\infty}\sum_{m=-\infty}^{\infty}C_n(a)C_m^*(a)e^{i(n-m)\Omega t}\times H_{nm}(\Delta\omega). \quad (3)$$

The parameter $\Delta\omega=\omega_a-\omega_s$ is written out explicitly for later application of the theory to experiments where only the time average of the transmission intensity is measured. The functions $H_{nm}(\Delta\omega)$ contain information on the line shape and hyperfine interactions. Their mathematical form resembles that of the transmission integral and the expressions are given elsewhere for single-line[15] and multiline[16] absorbers. In the limit of a thin single-line absorber,[13]

$$H_{nm}(\Delta\omega)\propto(\Delta\omega-n\Omega+i\Gamma)^{-1}-(\Delta\omega-m\Omega-i\Gamma)^{-1}, \quad (4)$$

where 2Γ is the linewidth (FWHM). The generalization to several modulated components is made by summation of appropriately weighted terms of the form of the right-hand side of Eq. (3).

To test the validity of Eq. (3) for magnetic modulation, experiments were performed with the Mössbauer resonance of ^{67}Zn. The source was a ZnO:^{67}Ga single crystal and the absorber was made of isotopically enriched ^{67}ZnO powder. The source was fixed to a support in the center of a superconducting solenoid producing an alternating magnetic field (Fig. 1). The magnetic field and the direction of observation of the gamma rays were perpendicular to the c axis of the single crystal.

Figure 2 shows the time dependence of the transmission intensity over the period of the magnetic field, oscillating sinusoidally at (a) 130 Hz, (b) 3.1 kHz, and (c) 9.3 kHz. The amplitude is $B_0=13.4$ mT in all cases. At low frequencies the magnetic sweep reveals a nearly Lorentzian shape [Fig. 2(a)]. As the modulation frequency approaches the linewidth of the ^{67}Zn resonance, time-dependent transient phenomena develop [Figs. 2(b) and 2(c)], resembling results reported for mechanical phase modulation.[14,15] Already at 3.1 kHz the line shape is considerably asymmetric due to the fact that the resonance is passed in a time comparable to the lifetime of the excited nuclear state. At 9.3 kHz, the asymmetry is even more pronounced.

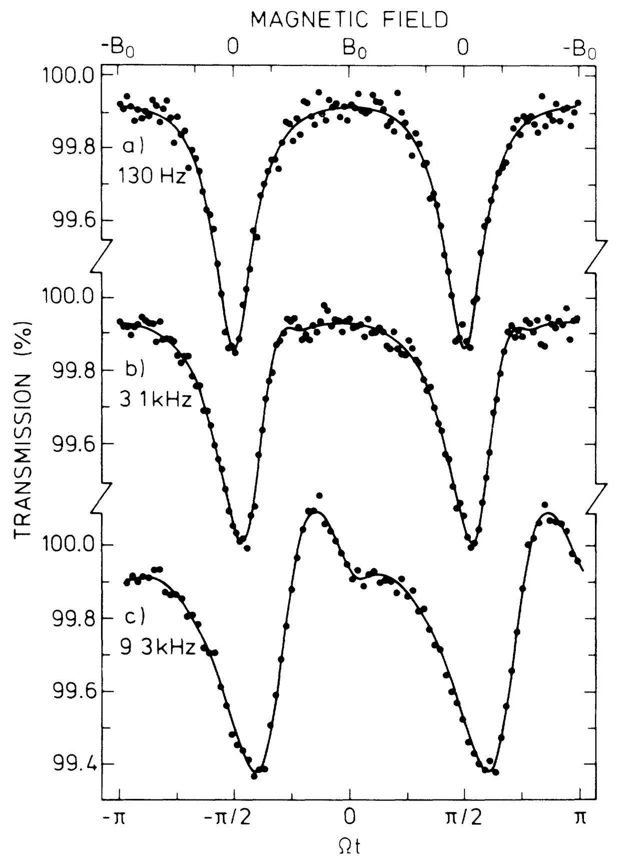

FIG. 2. Unfolded data from ^{67}Zn Mössbauer experiments with sinusoidal magnetic scanning. Time dependence of the transmission intensity is shown at three frequencies of the externally applied magnetic field with an amplitude of $B_0=13.4$ mT. Phase angle Ωt runs over the period of the modulation cycle.

The transmission curve in Fig. 3, measured at 6.2 kHz with $B_0=26.7$ mT, distinctly shows the new features. The counting rate after the resonance momentarily exceeds the nonresonant background level, unambiguously determined by the data. This phenomenon is a characteristic prediction of the present theory and it is caused by phasing of the contributions of the source and the absorber in the transmitted radiation field. The peak-to-peak effect observed is 0.9%, well above what

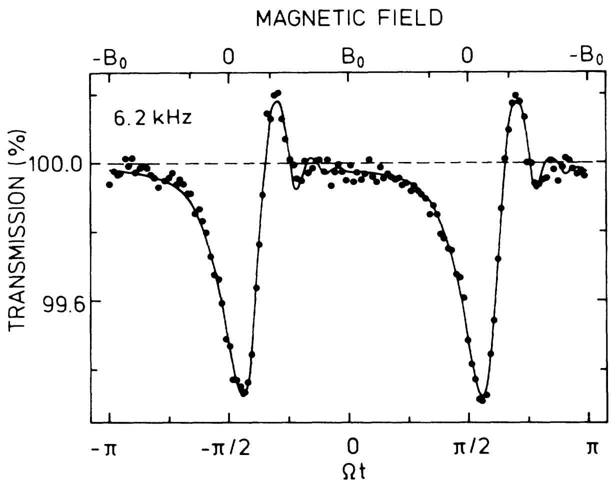

FIG. 3. Time dependence of the transmission intensity, measured with a magnetic field oscillating sinusoidally at 6.2 kHz with an amplitude of $B_0=26.7$ mT. The dashed line indicates the background intensity that would be observed in the absence of the Mössbauer resonance.

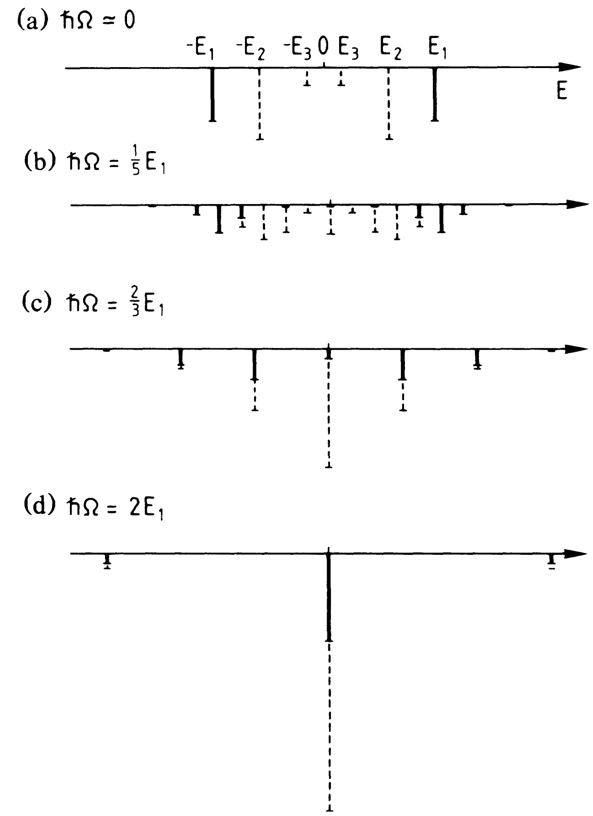

FIG. 4. Schematic ^{57}Fe Mössbauer spectra in a periodic magnetic field switching at the rate $\Omega/2\pi$ between two opposite directions. The contributions to the spectra originating from the resonance lines at $\pm E_1$ ($\pm E_2$, $\pm E_3$) are shown by thick bars (dashed lines).

was achieved in conventional ^{67}Zn experiments.

With perpendicular orientation of the c axis of the ZnO crystal relative to $\mathbf{B}(t)$ and the direction of observation of the gamma rays, 83% of the source radiation shows the Zeeman splitting of the 93-keV excited state only. Equation (1) describes the phase modulation of this component. Because of the large electric quadrupole interaction, the ground-state ($I_g=\frac{5}{2}$) energy levels $m_g=\pm\frac{5}{2}$ and $m_g=\pm\frac{3}{2}$ remain practically unsplit. The solid lines in Figs. 2 and 3 are least-squares fits according to a sum of two terms of the form of Eq. (3). The second, smaller component is influenced also by magnetic splitting of the substate $m_g=\pm\frac{1}{2}$. The theoretical curves accurately follow the characteristic features of the experimental data.

The data from transient experiments can be used to determine the modulation amplitude a with high accuracy.[14,15] This self-calibrating feature is utilized in a new type of measurement of the magnetic moment μ_e of the 93-keV state [see Eq. (1)], which offers also a sensitive test of the theory. The values obtained from the fits of Figs. 2(c) and 3 are $\mu_e=0.573(15)\mu_N$ and $0.595(13)\mu_N$, respectively. These values are in good agreement both with the earlier result[17] $\mu_e=0.58(3)\mu_N$ and with a value $\mu_e=0.589(9)\mu_N$, determined from our low-frequency experiments. Also other parameters obtained from the fits of the transient curves are in accordance with the values determined from auxiliary measurements. The agreement further confirms that the underlying theory is well justified in the case of magnetic modulation.

Starting from Eq. (3) it is straightforward to obtain predictions for results of *velocity domain* experiments in which the time average of the transmission intensity is measured. Integration over time gives

$$\bar{I}(\Delta\omega)=\frac{1}{T}\int_0^T I(\tau;\Delta\omega)d\tau = \sum_{n=-\infty}^{\infty}|C_n(a)|^2 W(\Delta\omega-n\Omega), \quad (5)$$

where $W(\Delta\omega-n\Omega)=H_{nn}(\Delta\omega)$ describes the basic line shape as a function of slowly changing $\Delta\omega$. Lorentzian shape is implied by Eq. (4). The sidebands are separated by the modulation frequency Ω and their relative intensity is given by $|C_n(a)|^2$.

As a specific example we consider the following model for ^{57}Fe: The static line positions in a magnetically split spectrum are denoted by $\pm E_1$, $\pm E_2$, and $\pm E_3$ [Fig. 4(a)], and the magnetic field seen by the nucleus is assumed to be switching periodically between two opposite directions. For each pair of lines $j=1,2,3$, the instantaneous frequency of the radiation field then switches between two values. A triangular phase-modulation function $\phi_j(t)$ with amplitude $a_j=\pi E_j/(2\hbar\Omega)$ is obtained by integration of the time-dependent Zeeman energy. With use of Eq. (2), the sideband intensities

$$|C_n(a_j)|^2 = [4a_j/(n^2\pi^2 - 4a_j^2)]^2 \sin^2 a_j \tag{6}$$

result for even n. For odd indices the sine function is replaced by cosine. Direct superposition of the subspectra with $j=1,2,3$ is strictly valid only for magnetic modulation in the source or in a thin absorber.

The above calculations can be applied to modeling of the rf-induced collapse phenomenon in iron-containing ferromagnetic materials. Magnetic materials showing the Mössbauer effect with ^{67}Zn are not known. The magnetization and the magnetic hyperfine field are assumed to be switching between two opposite directions at the rate determined by the frequency of the applied field. This may be close to the actual situation in magnetically soft materials. The model should show the main features related to the purely magnetically induced sidebands.

The dependence of the ^{57}Fe Mössbauer spectrum on the modulation frequency is illustrated in Fig. 4. At very low frequencies only the resonance lines corresponding to the static magnetic field are seen. When the modulation frequency is increased, clusters of sidebands appear around the original positions of these lines as indicated by thick bars for the outermost pair of the resonance lines [Fig. 4(b)]. At still higher frequencies the intensity collapses to the center line [Fig. 4(d)].

The observation of rf-induced collapse of the static hyperfine pattern indicates that the nuclei see a rapidly varying magnetic field. The appearance of the sidebands then requires only that the magnetic hyperfine field alternates periodically, a conclusion supported also by experiments[18] on the ^{181}Ta resonance with a nonmagnetic sample. Our model is qualitatively different from stochastic theories of domain-wall motion[3] and paramagnetic relaxation in which well-defined sidebands do not exist. However, at the limit of very high frequencies the intensity outside the collapsed center line approaches zero as $1/\Omega^2$, resembling the case of fast relaxation.

To summarize, the influence of the dynamic Zeeman effect on nuclear transitions is described classically as phase modulation of the emitted gamma radiation. Through ^{67}Zn Mössbauer measurements in the time domain, we provide unambiguous experimental evidence on the phase-modulation theory. The theory also predicts sidebands and a collapse phenomenon due to the dynamic Zeeman effect in magnetically soft materials exposed to rf fields. Magnetic transient experiments are used for a new type of determination of the magnetic moment of the 93-keV excited state of ^{67}Zn.

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